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Supporting Information

## **A Novel Graphene Quantum Dot-Based mRNA Delivery Platform**

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# Supplementary Materials

## Contents

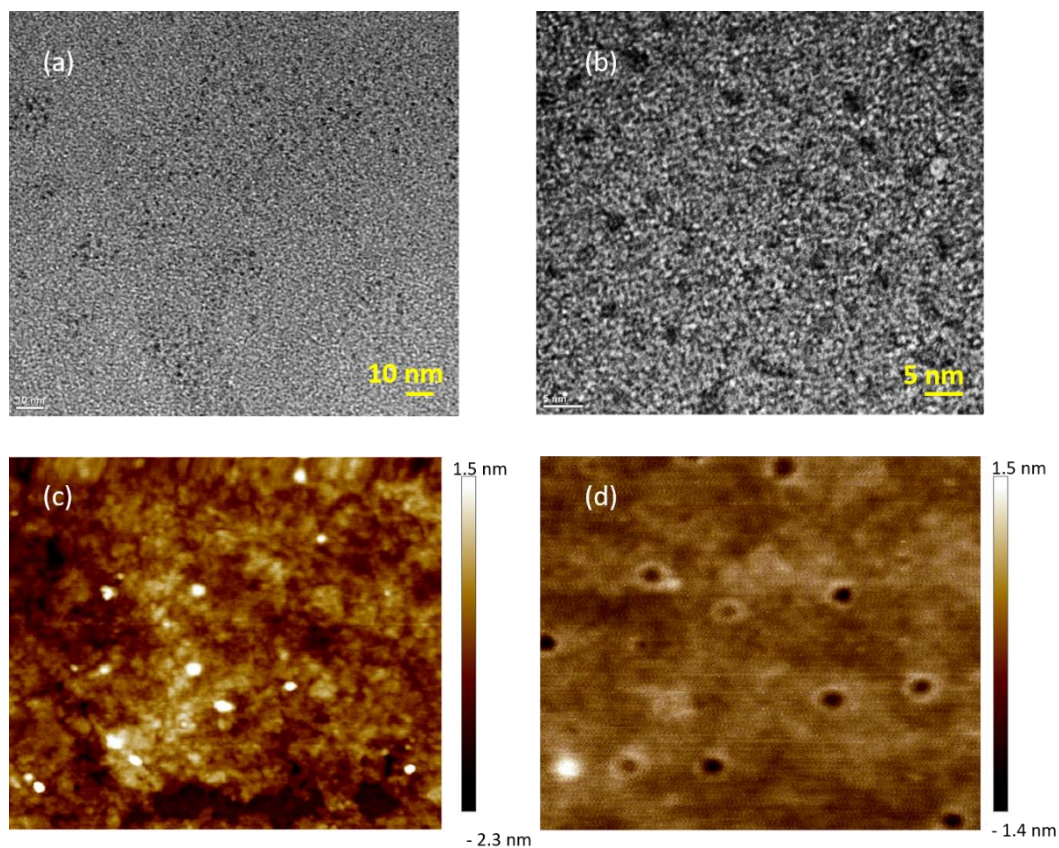
**Figure S1** (a) Low and (b) high resolution TEM images of GQDs; High resolution AFM images of (c) GQDs and (d) FGQDs.

**Figure S2** Surface zeta potential of FGQDs with pH value 4 and 14.

**Figure S3** Emission and UV–Vis absorption spectra of the GQDs and FGQDs.

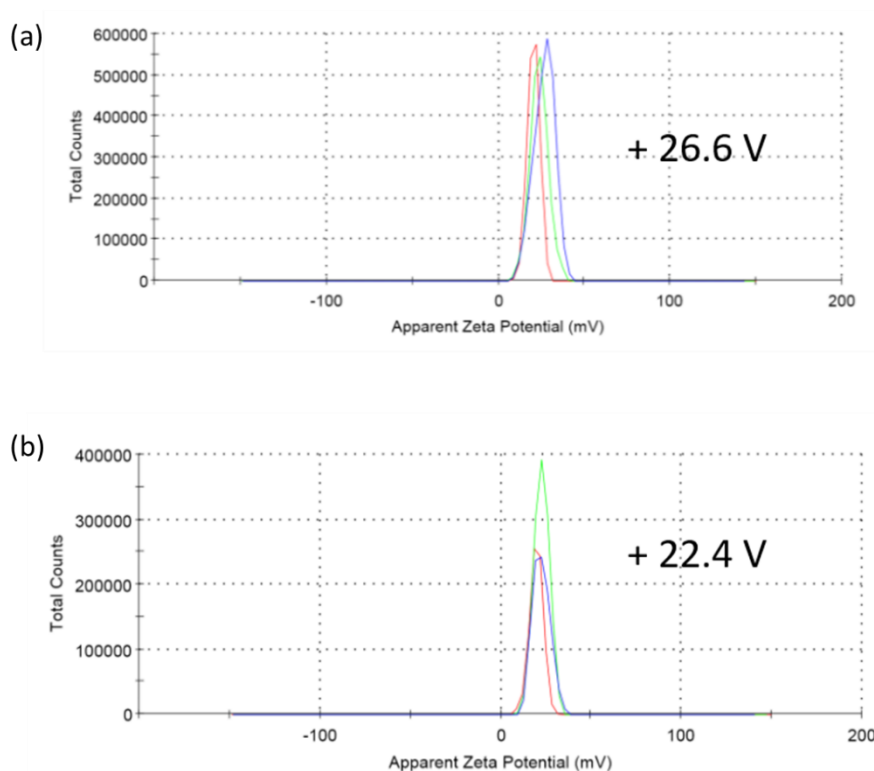
**Figure S4** Cell health data obtained following transfection.

**Table S1** FGQDs/mRNA doses for cell incubation.



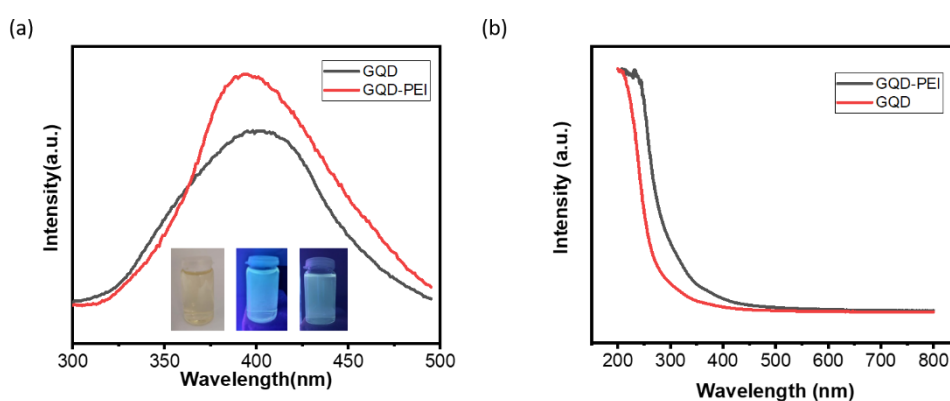
**Figure S1** (a) Low and (b) high resolution TEM images of GQDs; High resolution AFM images of (c) GQDs and (d) FGQDs.

As shown in Figure S1, the average geometry of GQDs is 2-3 nm. While after surface modification by PEI, FGQDs grows to tens of nanometers. The thickness of GQDs is less than 1 nm, indicating the GQDs were composed of 1-3 layers of graphene sheets. After functionalized, 4 layers of graphene sheets were observed in FGQDs with a thickness of 1.32 nm.



**Figure S2** Surface zeta potential of FGQDs under pH value as (a) 4 and (b) 14.

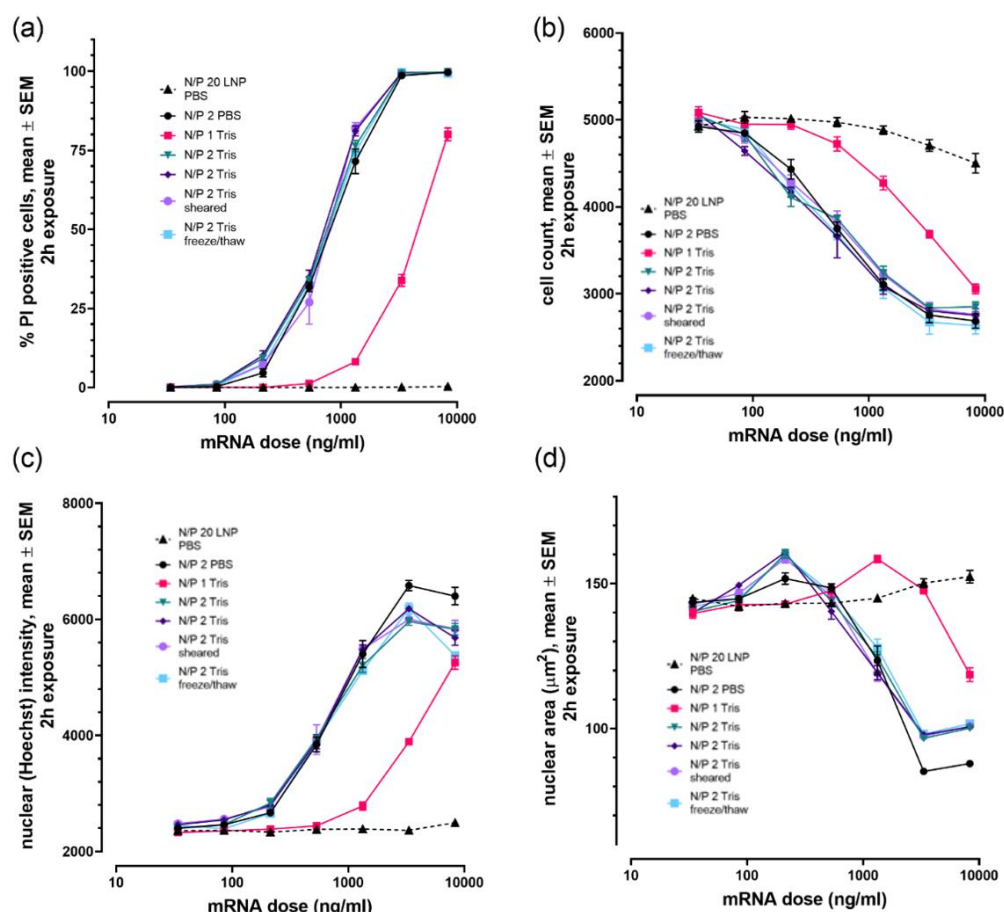
As shown in Figure S2, surface zeta potential of FGQDs keep positive at full range of pH value from 4 to 14. Such stability of zeta potential endows the prepared FGQDs to be widely used as platform for pH sensitive drugs.



**Figure S3** (a) Emission spectra of the GQD and FGQDs with excitation of 360 nm, the inset is photographs of the solution of GQD taken under visiblelight (left) and under 365 nm UV light (middle), and FGQDs under 365 nm UV light (Right); (b) UV–Vis absorption spectra of GQDs and FGQDs.

Figure S3 shows the UV absorption and fluorescence properties of GQDs and FGQDs. As shown in Fig. S3 (a), both GQDs and FGQDs are able to absorb UV light at 360 nm to emit strong green

fluorescence. Such property provide opportunity for GQD and FGQDs to be used to track drug release performance. Compared with GQDs, en enhanced UV-Vis absorption of FGQDs are observed in Figure S3 (b). No apparent absorption peak is observed in Fig S2, indicating homogenous geometry of GQDs and FGQDs.



**Figure S4.** (a) The percentage of propidium iodide positive cells following 2 h pulsed dose; (b) The number of positive cells following 24 h pulsed dose; (c) The nuclear intensity after 2 h exposure; (d) The nuclear area after 2 h exposure.

Figure S3 presents Cell health data obtained following transfection. The percentage of propidium iodide positive cells following 2 hours pulsed dose is shown in panel (a) for reference (this is also shown in Figure 6). Cellular responses to graphene-based delivery systems include nuclear swelling and increases in intensity, followed by continued increases in intensity and nuclear contraction. Curves show responses to FGQDs or reference LNPs complexed with mRNA at different charge ratios (N/P ratios).

Data is shown as mean  $\pm$  SEM from 6 wells for each point (combined data from 3 replicates in two separate experiments for all data points).

**Table S1** FGQDs/mRNA doses for cell incubation

ng/ well	ng/mL
250	8333
100	3333
40	1333
16	533
6.4	213
2.6	85
1.0	34
0	0